AD 660655

USNRDL-TR-67-116
2 July 1967

FRACTIONATION VERSUS PARTICLE TYPE IN NUCLEAR SURFACE SHOT SMALL BOY. Differences in Radiochemical Composition Between Fritted and Spheroidal Particles.

by

J. N. Pascual
E. C. Freiling

U.S. NAVAL RADIOLOGICAL DEFENSE LABORATORY

SAN FRANCISCO · CALIFORNIA · 94135

20

PHYSICAL CHEMISTRY BRANCH
E. C. Freiling, Head

NUCLEAR TECHNOLOGY DIVISION
R. Cole, Head

ADMINISTRATIVE INFORMATION

The work reported was part of a project sponsored by the Defense Atomic Support Agency under NWER Program A-7c, Subtask 10.052.

DDC AVAILABILITY NOTICE

Eugene P. Cooper

Eugene P. Cooper
Technical Director

D. C. Campbell, CAPT USN
Commanding Officer and Director

ABSTRACT

Spheroidal and fritted particles of three size classes from close-in fallout samples collected from Small Boy were separated by hand. Analysis of spheroidal and fritted groups from the > 1400-μ and the 350-700-μ size classes showed that spheroidal particles had higher specific activities than the fritted particles; however this difference was less pronounced for the smaller size groups. Contrary to reported observations on debris from a high-yield coral-surface burst, all groups showed comparable degrees of fractionation for radionuclides with rare-gas precursors. The Small Boy shot conditions were unique, however, and this homogeneity may not occur in more typical silicate-surface bursts. Ru^{106} and Sb^{125} were alike in behavior, but the data for these radionuclides do not correlate like those for rare-gas descendants. The necessity of adopting multidimensional correlations remains to be evaluated.

SUMMARY

The Problem

During the field phase of Shot Small Boy, an attempt was made to determine the effect of particle type on radionuclide fractionation. Because of suspected errors in sample designation, the results of that study could never be utilized.

Findings

Gross samples from Shot Small Boy were separated according to particle size by gentle hand sieving. Samples of fritted and spheroidal particles were hand-picked from the larger sized fractions. These fractions were studied for their physical and radiochemical properties, and the results were used to determine the importance of radionuclide fractionation with particle type from an operational viewpoint.

INTRODUCTION

It has long been recognized that several different types of particles could be distinguished in fallout from land surface bursts.[1] In 1958, Mackin et al. reported the radiochemical compositions of spherical and irregular particles observed in debris from a high-yield coral-surface burst.[2] They found the spherical particles to be highly depleted in fission-product mass chains with rare-gas precursors, while the irregular particles were relatively representative. R. Heft has recently found evidence for two families of particles in cratering bursts.* One family is crystalline and the other amorphous. Both of these families appear to have rare-gas precursor nuclides distributed according to the particle surface area, but only the amorphous particles contain refractorily behaving nuclides. From the viewpoint of fallout hazard prediction, it is important to determine if a similar effect exists in debris from silicate-surface bursts, and if so, whether the magnitude of the effect is such as to require inclusion in fallout prediction systems.

Shot Small Boy in 1962 offered an opportunity for study of the effects of fractionation with particle type.[3] Consequently, during the field phase of that operation, personnel at the Nevada Test Site made a special effort to obtain an early recovery (ER) sample and to separate, by hand, groups of spherical and irregular particles. A gross sample, two separated groups, and the residual sample, were all returned to the U. S. Naval Radiological Defense Laboratory (NRDL) for additional documentation and analysis. After documentation the samples were sent to a commercial laboratory for radiochemical analysis. Table 1 combines the results obtained from the two analyses and incorporates a switch of sample designation (ER-2 and ER-5) suggested by field personnel.

The data in this table do not make sense. If we first compare the equivalent fissions in the samples as determined by Zr^{95} and by Mo^{99}, and as estimated by ionization, we see that there is good agreement between these three values for samples ER-2 and ER-6 but poor agreement for samples ER-4 and ER-5. In spite of this, the sample weights appear

*R. Heft, Lawrence Radiation Laboratory (Livermore), private communication.

TABLE 1

Data on Early Recovery Samples

	ER-2 Residual Sand all spheres and fritted particles removed	ER-4 Spherical Particles picked from fallout material	ER-5 Gross Sample including spheres and fritted particles	ER-6 Fritted Particles picked from fall-out material
Weight (mg)	409.0	10.1	72.0	57.6
Equivalent fissions of Zr^{95}	6.5×10^{12}	(3.8×10^{11})a	(5.7×10^{11})	3.8×10^{13}
Equivalent fissions of Mo^{99}	5.9×10^{12}	(7.2×10^{11})	(1.1×10^{12})	3.6×10^{13}
Fissions estimated by ionization	9.0×10^{12}	1.1×10^{13}	1.7×10^{13}	2.2×10^{13}
Estimated fissions per gram (ionization)	2.2×10^{13}	1.1×10^{15}	2.4×10^{14}	3.8×10^{14}
$r_{1,95}$ values for fission products:				
Sr^{89}	0.0104	(2.41)	(1.90)	0.0121
Sr^{90}	(0.0374)	(3.04)	(3.48)	0.378
Y^{91}	-	-	-	0.0502
Te^{132}	-	-	-	0.0467
Cs^{136}	(0.0536)	-	-	(0.456)
Cs^{137}	(0.0273)	-	-	0.0762
Ba^{140}	(0.905)	-	-	(0.990)
Ce^{141}	-	-	-	(5.38)
Ce^{144}	-	-	-	(0.244)

a. Values in parentheses are not consistent either with other values in this table or with previously reported observations.

to be reasonable and the estimated fissions per gram appear to be in the right order. Thus, as would be expected, the residual sand has the lowest specific activity, the gross sample has the next lowest specific activity and the spherical particles have the highest specific activity. If we look now at the $r_{1,95}$ values for fission products, the spherical particles and gross samples are seen to be enriched in Sr^{89}. By comparison with the results of Mackin et al.,[2] we would expect the spherical-particle fraction to be the most depleted of all. As far as internal consistency with other Small Boy samples is concerned,[4] these values would make samples ER-4 and ER-5 more than 3 times richer in Sr^{89} than the next most highly enriched sample observed in the local fallout.

The present work is a report of an effort to resolve these anomalies. The effort was undertaken when the samples were 20 months old, and therefore only partial correspondence could be achieved between the nuclides determined in the old studies and in the new ones. Limited funding precluded the analysis of gross and residual samples.

BACKGROUND

Shot Small Boy was a low yield burst at the Nevada Test Site in 1962. The device was placed on a wooden tower, 10 feet above a dry lake bed, the material of which consisted of alluvial silt. Pre-shot sieve analysis showed that the soil contained no particles greater than 44 microns. The two fallout samples studied were designated S2-PC-22 and 305-AO-1. These samples were collected during the field phase of the operation at distances of 9,200 and 13,300 feet from ground zero, respectively. The large particles used in this study were formed by the coalescence of molten or partially molten soil during the high-temperature phase of the detonation.

EXPERIMENTAL

Size separation of the two fallout samples was obtained with a minimum of disturbance to the original states by gentle hand shaking of a nest of 4-in. diameter Tyler Standard Screen Scale sieves in a plastic gloved box as described in a report to be published.* Groups of spheroidal and fritted particles were selected by hand from the

*J. N. Pascual, "Bias in Fallout Data from Nuclear Surface Shot Small Boy. An Evaluation of Sample Perturbation by Sieve Sizing," USNRDL-TR.

larger size fractions. The particles chosen from the 350-700 micron fraction of sample 305-AO-1 are shown in Figs. 1 and 2. The particle type fractions were then documented for weight, gross gamma-ray activity, and gamma-ray spectra before transmittal to a commercial laboratory (Tracerlab, Inc. (TLW)) for radiochemical analysis of long-lived fission products. The fritted particles were found to be either dark-brown, white, or off-white, while the glassy spheroidal particles were light-green, brown or black.

RESULTS

Table 2 shows the weights and gross gamma-ray activities for the particle size and type groups. The gamma-ray activities are of interest only for comparison of particle activities and as a check on the radiochemical results. In each case the specific activity of the spheroidal particles is greater than that of the fritted particles in the same size class. However, the specific activity of the fritted particles increases as the size decreases, while in most instances the specific activity of the spheroidal particles decreases with decreasing size, so that the net result as size decreases is a convergence of specific activities for the two types of particles. The mass per particle for the spheroidal particles is sometimes greater than, and sometimes less than, that for the fritted particles of the same size. This probably reflects a variable vesicularity for the spheroidal particles.

Figure 3 shows the gamma-ray spectra of the particle groups from Sample 305-AO-1. These are the two extreme cases. The dashed line, for the large spheroidal particles, shows a pronounced depletion in the peaks for Ru^{106}-Rh^{106} and Sb^{125} relative to Ce^{144}-Pr^{144}.

Table 3 shows the results of the radiochemical analysis. These results support those from gross activity and from gamma-ray spectra. The equivalent fissions per gram are roughly proportional to the specific activities shown in Table 2. All samples show comparable depletion of Sr^{90} and Cs^{137}, while the Ru^{106} ratios and the Sb^{125} ratios both vary over a factor of 10. These results correspond to the data from the gamma-ray spectra.

NRDL 506-67

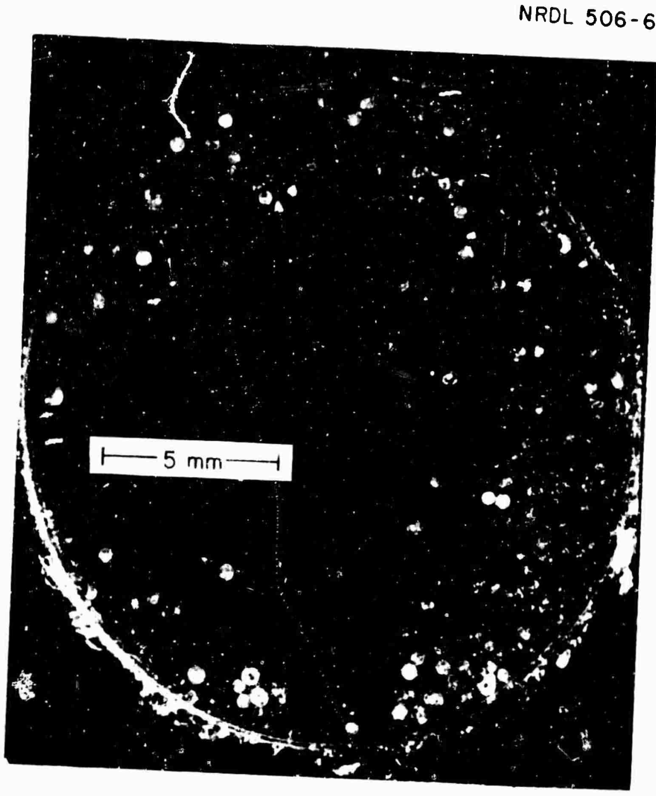

Fig. 1 Spheroidal Particles (Group 3S) from Sample 305-A0-1.

NRDL 506-67

Fig. 2 Fritted Particles (Group 3F) from Sample 305-AO-1.

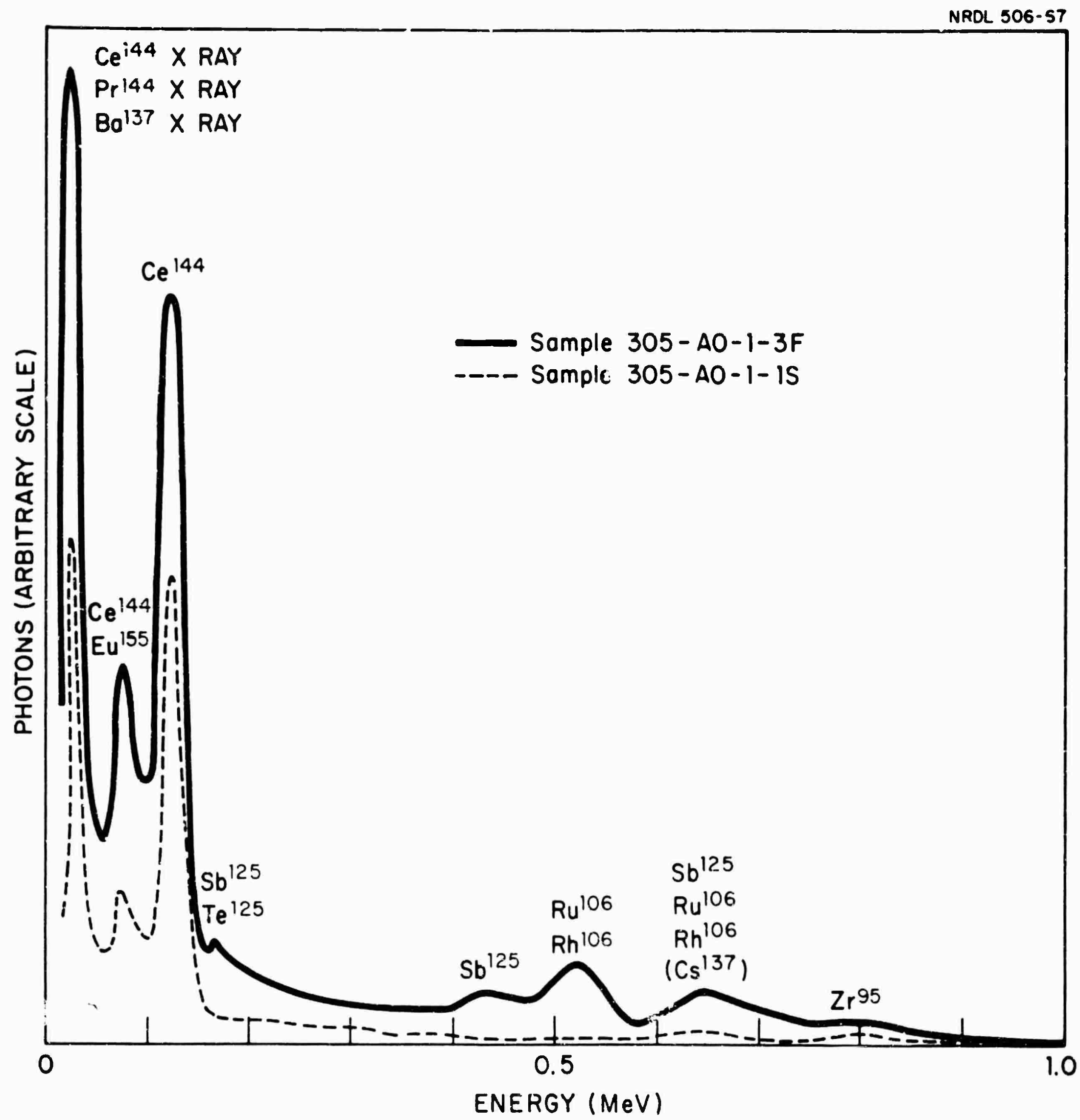

Fig. 3 Gamma-ray Spectra for Particle Groups 1S and 3F of Sample 305-AO-1. In 1S, Sb^{125} and Ru^{106} are seen to be depleted with respect to Zr^{95} and Ce^{144} by a factor of 10 as compared to 3F.

TABLE 2

Mass and Gross-Activity Data for Picked Particles

Fraction	Particle Size (μ)	Particle Type	Number of Particles	Total Mass (mg)	Total Activity (10^4 cpm)	Specific Activity (cpm/mg)	Mass per Particle (mg)	Activity per Particle (10^4 cpm)
			A. Sample S2-PC-2					
1	> 1400	Spheroidal	2	13.7	12.8	9340	6.8	6.4
		Fritted	4	14.5	8.34	5750	3.6	2.1
2	700-1400	Spheroidal	11	19.3	13.6	7020	1.75	1.2
		Fritted	20	17.0	9.8	5800	0.85	0.5
			B. Sample 305-AO-1					
1	> 1400	Spheroidal	6	29.5	5.05	1710	7.4	1.3
		Fritted	10	80.3	0.16	20	8.0	0.016
2	700-1400	Spheroidal	68	61.7	33.4	5460	0.91	0.49
		Fritted	85	114.7	27.3	2070	1.4	0.28
3	350-700	Spheroidal	300	76.6	51.3	6700	0.25	0.17
		Fritted	331	35.0	11.3	3240	0.11	0.034

TABLE 3

Data on Sample 305-AO-1

Sample	1F	1S	3F	3S
Equivalent fissions of Ce^{144} per gram	4.98×10^{12}	4.08×10^{14}	6.40×10^{14}	1.44×10^{15}
$r_{1,144}$ values for fission products:				
Sr^{90}	0.22	0.312	0.259	0.248
Ru^{106}	0.0677	0.0194	0.228	0.0958
Sb^{125}	0.0369	0.0104	0.120	0.0823
Cs^{137}	0.0596	0.0345	0.0750	0.0517

DISCUSSION

Comparison of the data in Table 3 with that of Table 1 requires the assumption that Ce^{144} did not fractionate from Zr^{95}. This appears reasonable in view of the low correlation parameter of 0.03 for $r_{144,95}$ reported by Crocker, et al.[5,*] On this basis, the new data confirm the estimated-fissions-per-gram values and the Cs^{137} values of Table 1. The Sr^{90} value for ER-6 also appears to be correct. Correction of the Sr^{89} values by use of the estimated fissions gives 0.083 and 0.064 for ER-4 and ER-5 respectively. Correction of Sr^{90} values gives 0.105 and 0.117 for ER-4 and ER-5, respectively. All four corrected values appear reasonable. They also move the points for these samples to reasonable positions in Crocker's correlation diagram.[4] However, there are so many anomalies in the remaining data that it appears risky to accept any of it.

The new data indicate that Cs^{137}, Sb^{125} and Ru^{106} all have about the same degree of volatility. Crocker et al., reported fractionation correlation values of 1.19 for Cs^{137} and 0.89 for Ru^{106} in Small Boy with reservations about the latter value.[5] The new data show these reservations to be well grounded because neither the Ru^{106} nor the Sb^{125} data correlate linearly with the data for nuclides with gaseous precursors. For Cs^{137} and Sr^{90}, differences in $r_{i,144}$ values shown by the new data here are not very dramatic. In general, the spheroidal particles, which were subjected to high temperatures, are somewhat more depleted in volatiles than the fritted particles (subjected to lower temperatures) of the same size. The depletion of larger particles with respect to smaller particles of the same type is actually more pronounced. These data therefore do not offer any evidence for two families of particles in this size range for this shot. This does not preclude the possibility of such an effect's being observed in other size classes. Because the burst height and pre-shot soil size for Small Boy were unique, it does not preclude the possibility for other silicate-surface bursts either. Our search for two families of particles in silicate-surface bursts is continuing under another project.

---

*This low correlation parameter indicates that the ratio of Ce^{144} to Zr^{95} is virtually independent of the degree of fractionation $r_{89,95}$.

REFERENCES

1. C. E. Adams, N. H. Farlow and W. R. Schell, Geochim. Cosmochim. Acta 18:42 (1960).

2. J. L. Mackin, P. E. Zigman, D. L. Love, D. Macdonald and D. Sam, J. Inorg. Nucl. Chem. 15:20 (1960).

3. E. C. Freiling, L. R. Bunney and F. K. Kawahara, "Operation SUN BEAM, Shot Small Boy, Proj.2.10,Physicochemical and Radiochemical Analysis (U)," Defense Atomic Support Agency POR-2216, U.S. Naval Radiological Defense Laboratory, 15 October 1964 (Classified).

4. G. R. Crocker, "The Effect of Radionuclide Fractionation on the Normalization Factor for Fallout Fields," U. S. Naval Radiological Defense Laboratory, USNRDL-TR-892, 4 August 1965 (see Fig. 1).

5. G. R. Crocker, F. K. Kawahara and E. C. Freiling, "Radiochemical-Data Correlations on Debris from Silicate Bursts," in Radioactive Fallout from Nuclear Weapons Tests, U. S. Atomic Energy Commission, Division of Technical Information, November 1965.

UNCLASSIFIED
Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author) U. S. Naval Radiological Defense Laboratory San Francisco, California 94135	2a. REPORT SECURITY CLASSIFICATION UNCLASSIFIED 2b. GROUP

3. REPORT TITLE

FRACTIONATION VERSUS PARTICLE TYPE IN NUCLEAR SURFACE SHOT SMALL BOY. Differences in Radiochemical Composition Between Fritted and Spheroidal Particles

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

5. AUTHOR(S) (First name, middle initial, last name)

Juan N. Pascual
Edward C. Freiling

6. REPORT DATE 3 November 1967	7a. TOTAL NO. OF PAGES 22	7b. NO. OF REFS 5

8a. CONTRACT OR GRANT NO. b. PROJECT NO. DASA, NWER Program A-7c, Subtask 10.052 c. d.	9a. ORIGINATOR'S REPORT NUMBER(S) USNRDL-TR-67-110 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)

10. DISTRIBUTION STATEMENT

This document has been approved for public release and sale; its distribution is unlimited.

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY Defense Atomic Support Agency Washington, D. C. 20301

13. ABSTRACT

Spheroidal and fritted particles of three size classes from close-in fallout samples collected from Small Boy were separated by hand. Analysis of spheroidal and fritted groups from the >1400-μ and the 350-700-μ size classes showed that spheroidal particles had higher specific activities than the fritted particles; however this difference was less pronounced for the smaller size groups. Contrary to reported observations on debris from a high-yield coral-surface burst, all groups showed comparable degrees of fractionation for radionuclides with rare-gas precursors. The Small Boy shot conditions were unique, however, and this homogeneity may not occur in more typical silicate-surface bursts. Ru^{106} and Sb^{125} were alike in behavior, but the data for these radionuclides do not correlate like those for rare-gas descendants. The necessity of adopting multidimensional correlations remains to be evaluated.

DD FORM 1 NOV 65 1473 (PAGE 1)
S/N 0101-807-6801

UNCLASSIFIED
Security Classification

14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Fractionation						
Fallout						
Particles						
Nuclear debris						
Fission products						